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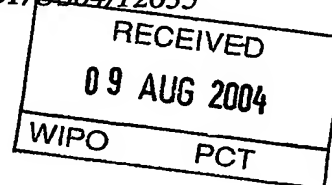
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PROVISIONAL APPLICATION FOR PATENT COVER SHEET
This is a request for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

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INVENTOR(S)

Given Name (first and middle [if any])	Family Name or Surname	Residence (City and either State or Foreign Country)
Benedetto Anthony	Iacovelli	Asheville, North Carolina

☐ Additional inventors are being named on the _____ separately numbered sheets attached hereto

TITLE OF THE INVENTION (500 characters max)

IMPROVED DESIGNS FOR A REDUCED COST, FLEXIBLE FUEL, METHANOL CAPABLE FUEL CELL AND COMPONENTS

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ENCLOSED APPLICATION PARTS (check all that apply)

☑ Specification *Number of Pages* 31
☑ Drawing(s) *Number of Sheets* 14
☐ Application Data Sheet. See 37 CFR 1.76

☐ CD(s), Number
☐ Other (specify)

METHOD OF PAYMENT OF FILING FEES FOR THIS PROVISIONAL APPLICATION FOR PATENT

☑ Applicant claims small entity status. See 37 CFR 1.27.
☑ A check or money order is enclosed to cover the filing fees
☐ The Commissioner is hereby authorized to charge filing fees or credit any overpayment to Deposit Account Number:
☐ Payment by credit card. Form PTO-2038 is attached.

FILING FEE AMOUNT ($)

$80.00

The invention was made by an agency of the United States Government or under a contract with an agency of the United States Government.
☑ No.
☐ Yes, the name of the U.S. Government agency and the Government contract number are: _____

Respectfully submitted,

SIGNATURE _____ Date | 04/22/2003

TYPED or PRINTED NAME ___ David M. Carter

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REGISTRATION NO. (if appropriate) | 26,407
Docket Number: | IAC-1

USE ONLY FOR FILING A PROVISIONAL APPLICATION FOR PATENT

PROVISIONAL UNITED STATES PATENT APPLICATION
35 U.S.C. § 111(b)

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Title: IMPROVED DESIGNS FOR A REDUCED COST,
FLEXIBLE FUEL, METHANOL CAPABLE FUEL
CELL AND COMPONENTS

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Background of the invention

Fuel cell technology has extraordinary potential, not just economically, but also to supply our power and transportation needs while drastically reducing global pollution. However, it is a nascent technology with several serious obstacles yet to overcome that preclude extensive market viability. The first obstacle is cost. While the materials and manufacturing costs are dropping at a rapid clip, most fuel cell technologies are, as yet, three to ten times more expensive than the technologies with which they compete. Cost has kept fuel cells to niche, "low hanging fruit" applications so far. The second obstacle is sensitivity to contaminants, which poison the electrodes and degrade the electrolytes, seriously limiting cell lifespan. A further problem is the reliance on expensive and esoteric materials. The low temperature fuel cells require expensive and rare catalysts; the high temperature fuel cells require esoteric materials to withstand the heat and caustic internal environments. The low temperature fuel cells are also very sensitive to poisoning from substances common in the existing fuel stocks.

Another obstacle facing fuel cells is that while the fuel cell stack has evolved into a simple compact design, it relies on extensive, costly supporting apparatus termed the 'balance of plant.' The balance of plant equipment is several times larger and often much more expensive than the fuel cell stack itself. For most applications, it is the expense of the balance of plant apparatus that prices the unit outside of commercial viability.

The last major problem plaguing the industry is hydrogen. Hydrogen, a chemically ideal fuel, is prohibitively expensive and dangerous, and lacks the infrastructure for broad use. Hydrogen makes metal brittle, escapes through microscopic cracks, and ignites with even a static electricity spark. In order to contain a useful amount of hydrogen, bulky, expensive hydride tanks, expensive cooling machinery, or high pressures are needed, all of which more than offset hydrogen's advantages.

Hydrogen fueled PEM technology currently leads the industry. It has many important advantages, but is not without multiple weaknesses. An in depth analysis of the challenges facing PEM technology can be found in appendix A.

A fuel cell that can run directly on methanol is the Holy Grail for the industry. Methanol is a fairly ideal fuel. It is colorless, odorless, energy dense liquid. Unlike hydrogen, methanol is easy to manufacture, store and transport with the existing infrastructures. It is inexpensive and does not need to be cooled or pressurized. It is available ubiquitously and can be manufactured locally form agricultural waste. Methanol has the added benefits of being a sustainable and environmentally friendly fuel source. Methanol has virtually none of the storage dangers of hydrogen, and there already is an existing infrastructure that can distribute it.

Unfortunately, as yet, the direct methanol fuel cell technology has not achieved commercial viability. The current art has serious 'balance of plant', efficiency, cost, crossover, poison sensitivity, and longevity problems. They don't use the fuel efficiently and require greater amounts of precious metal catalysts. The improvements described herein solve many of the hurdles facing direct methanol and many other types of fuel cells.

Objectives

I have invented a family of improved fuel cell components and processes that allow for the manufacture of improved fuel cells. The parts can be used to enhance other fuel cell

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designs, or collectively in a new and vastly improved fuel cell system. These designs lend themselves especially to direct methanol or borohydrate powered fuel cell applications, but can be utilized with a broad range of fuel stocks.

Specifically, the objectives of these designs are to provide improved fuel cells, components and processes that, compared to the current state of the art, are:

> Much less expensive to build.
> Longer lasting.
> Much more efficient, powerful, and have a much higher energy density.
> Able to operate using on a variety of inexpensive and readily available fuels, including methanol.
> Able to eliminate the sensitivity to poisoning by carbon monoxide and sulfur compounds that currently severely limits the lifespan, feasibility, and commercial viability of the current art fuel cells.
> Able to use little or no platinum with the ability to employ multiple types of advantageous catalysts.
> Able to function with much less balance of plant apparatus.
> Able to utilize catalysts more efficiently, decreasing necessary catalyst loadings especially to achieve or surpass the goals set forth by the US department of energy for fuel cell loading.
> Able to incorporate a means of reducing or eliminating electrolyte degradation.
> Less sensitive to vibration and wear.
> Able to eliminate water balance issues completely
> Part of a rechargeable fuel cell.
> Capable of electric and/or magnetic field augmentation of mass transport and diffusion as well as advantageous charge bilayer manipulation.
> Ready for broad commercialization.

Advantages

This new technology enables the creation of a new type of fuel cell that does not have the barriers to broad commercialization that hold back current art. These designs could <u>more than halve the cost</u> of fuel cells. The components can use multiple fuels, especially methanol and sodium tetrahydroborate. They last much longer than current art and are less sensitive to vibration and wear. They have smaller stacks with increased power densities and less balance of plant apparatus. They are less sensitive to ambient or operating conditions and are scalable to a broad range of applications. They should penetrate deeply into the largest range of fuel cell consumer markets.

Advantages Explained

Increased efficiency

These components and processes utilize catalysts and fuels more efficiently than the currently available designs.

- They have better substrate/catalyst interaction that greatly increases the reaction rate and current density.
- They enable faster mass transport.
- They eliminate concentration gradient efficiency losses.

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- Ohmic losses are reduced by an order of magnitude. The internal resistance and impedance are nearly insignificant, certainly much lower than in any commercially available fuel cells.
- The flow through operation allows for more efficient water handling.
- Ther\e is substantially reduced fuel crossover.
- They employ a novel, simple, self-regulating, nonparasitic means of thermal and water management.
- The flow-through design allows oscillating flow.
- They do not rely on triple phase interaction for the reaction to take place.

Another means for efficiency gains embodied in these designs is the ability to use a means to manipulate and optimize the charge bilayer and ionic flow conditions.

Less catalyst requirements

They provide greater overall catalyst surface area <u>and</u> greater catalyst surface area to mass ratio, decreasing necessary catalyst loading. Further, the design intrinsically eliminates many of the causes of decreased substrate/catalyst interaction. Therefore less catalyst and/or less expensive types of catalysts can be used, decreasing costs. They achieve and surpass the goals set forth by the US Department of Energy for fuel cell catalyst loading.

Lowered materials and manufacturing costs

The new electrode designs lend themselves to ease of inexpensive mass manufacturing. Depending on the manufacturing processes employed, they can be made on one machine and can bypass the limits of electro- and electroless plating.

Current designs for low temperature fuel cells employ field flow plates. Up to 80% of the volume of the fuel cell stack and a large percentage of the cost is for the field flow plates. The designs described below do not require field flow plates, further decreasing costs.

The increased electrode current density and novel means for eliminating field flow plates improves the energy density of the stack. This allows the manufacture of smaller less costly but just as powerful fuel cells, which in turn increases the range of applications.

Decreased balance of plant

Taking greater advantage of intrinsic synergies, this fuel cell can utilize much simpler water balance, thermal management, current collection, and electrolyte management systems. The cell will require up to 90% less physical balance of plant supporting apparatus, such as pumps, current collectors, heat exchangers, evaporators, belts, hoses, controllers, interconnects, wiring, etc...

Because the improved electrodes can be part of technology that enables fuel cell stacks to be built without the added cost and bulk of field flow plates and because less electrode area can be used, the stacks themselves should be significantly smaller than comparable current art devices.

Ultimately, these savings in equipment decrease the cost of materials and manufacturing, while eliminating much of the current art's parasitic energy drains.

There will be less required maintenance and less parasitic efficiency losses from balance of plant apparatus. Again this will increase efficiency and decrease cost.

FEE TRANSMITTAL
for FY 2003
Patent fees are subject to annual revision.

☑ Applicant claims small entity status. See 37 CFR 1.27

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Complete if Known

Application Number	
Filing Date	04/22/2003
First Named Inventor	Benedetto Anthony Iacovelli
Examiner Name	
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Attorney Docket No.	IAC-1

METHOD OF PAYMENT (check all that apply)

☑ Check ☐ Credit card ☐ Money Order ☐ Other ☐ None

☐ Deposit Account:

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The Commissioner is authorized to: *(check all that apply)*
☐ Charge fee(s) indicated below ☐ Credit any overpayments
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FEE CALCULATION

1. BASIC FILING FEE

Large Entity Fee Code	Fee ($)	Small Entity Fee Code	Fee ($)	Fee Description	Fee Paid
1001	750	2001	375	Utility filing fee	
1002	330	2002	165	Design filing fee	
1003	520	2003	260	Plant filing fee	
1004	750	2004	375	Reissue filing fee	
1005	160	2005	80	Provisional filing fee	80.00

SUBTOTAL (1) ($) 80.00

2. EXTRA CLAIM FEES FOR UTILITY AND REISSUE

	Extra Claims		Fee from below	Fee Paid
Total Claims [ ]	-20** = [ ]	X [ ]	= [ ]
Independent Claims [ ]	-3** = [ ]	X [ ]	= [ ]
Multiple Dependent		[ ]	= [ ]

Large Entity Fee Code	Fee ($)	Small Entity Fee Code	Fee ($)	Fee Description
1202	18	2202	9	Claims in excess of 20
1201	84	2201	42	Independent claims in excess of 3
1203	280	2203	140	Multiple dependent claim, if not paid
1204	84	2204	42	** Reissue independent claims over original patent
1205	18	2205	9	** Reissue claims in excess of 20 and over original patent

SUBTOTAL (2) ($)

**or number previously paid, if greater; For Reissues, see above*

FEE CALCULATION (continued)

3. ADDITIONAL FEES

Large Entity Fee Code	Fee ($)	Small Entity Fee Code	Fee ($)	Fee Description	Fee Paid
1051	130	2051	65	Surcharge - late filing fee or oath	
1052	50	2052	25	Surcharge - late provisional filing fee or cover sheet	
1053	130	1053	130	Non-English specification	
1812	2,520	1812	2,520	For filing a request for ex parte reexamination	
1804	920*	1804	920*	Requesting publication of SIR prior to Examiner action	
1805	1,840*	1805	1,840*	Requesting publication of SIR after Examiner action	
1251	110	2251	55	Extension for reply within first month	
1252	410	2252	205	Extension for reply within second month	
1253	930	2253	465	Extension for reply within third month	
1254	1,450	2254	725	Extension for reply within fourth month	
1255	1,970	2255	985	Extension for reply within fifth month	
1401	320	2401	160	Notice of Appeal	
1402	320	2402	160	Filing a brief in support of an appeal	
1403	280	2403	140	Request for oral hearing	
1451	1,510	1451	1,510	Petition to institute a public use proceeding	
1452	110	2452	55	Petition to revive - unavoidable	
1453	1,300	2453	650	Petition to revive - unintentional	
1501	1,300	2501	650	Utility issue fee (or reissue)	
1502	470	2502	235	Design issue fee	
1503	630	2503	315	Plant issue fee	
1460	130	1460	130	Petitions to the Commissioner	
1807	50	1807	50	Processing fee under 37 CFR 1.17(q)	
1806	180	1806	180	Submission of Information Disclosure Stmt	
8021	40	8021	40	Recording each patent assignment per property (times number of properties)	
1809	750	2809	375	Filing a submission after final rejection (37 CFR 1.129(a))	
1810	750	2810	375	For each additional invention to be examined (37 CFR 1.129(b))	
1801	750	2801	375	Request for Continued Examination (RCE)	
1802	900	1802	800	Request for expedited examination of a design application	

Other fee (specify) _____

*Reduced by Basic Filing Fee Paid

SUBTOTAL (3) ($)

SUBMITTED BY

Name (Print/Type)	David M. Carter	Registration No. (Attorney/Agent)	26,407	Telephone	(828) 252-6225
Signature	✗ [signature]			Date	04/22/2003

(Complete if applicable)

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Greater durability and longevity

Compared with the current art, these electrodes have much better structural integrity and are less sensitive to vibration, wear, and corrosion. They employ a means of eliminating the build up of the toxins that degrade the catalysts and shorten the cell life. They will also be refurbishable and recyclable by the manufacturer.

A permutation of these designs incorporates or allows for a means of reversing the natural degradation of the electrolyte that occurs over time, which will extend cell life.

Because they allow for designs with reduced balance of plant systems there is less equipment to break down. Simpler is better. These features confer markedly increased durability and longevity.

Better fuel selection

Low temperature fuel cells tend to use hydrogen as a fuel. Hydrogen is volatile, explosive, and expensive. It makes metals brittle and escapes trough microscopic cracks. Storage techniques are too bulky, expensive and/or risky. Further, there is no adequate infrastructure for hydrogen storage and distribution.

This fuel cell can run on a variety of fuels that do not have the above problems, including methanol, ethanol, and borates.

No water balance issues

Current fuel cells are very susceptible to water balance problems. They need to have a specific degree of water concentration at each part. If any part gets too dry or too wet it will malfunction. This problem is especially important because waste product water is formed at the cathode, which can flood it, and water is dragged from the anode drying it. Furthermore, fuel cells operate in changing ambient humidity and encounter frequent temperature fluctuations.

This creates a cell that is essentially never functioning optimally, is quite sensitive to ambient and operating conditions, and requires additional water handling equipment that parasitizes the efficiency.

The natural synergies of my new design crate a cell that is essentially impervious to the kinds of water balance problems that can paralyze the current art. They obviate the need for most water handling equipment.

Decreased sensitivity to contaminants.

These electrodes can incorporate a means to refresh themselves while inside the fuel cell to prevent poisoning by sulfur, carbon monoxide and the like.
Further, they can be factory refurbished or recycled for a still longer usable life span.

Applicable to a greater range of markets

Because they:
- Cost much less and last much longer
- Operate on multiple fuels especially methanol and sodium tetrahydroborate
- Are less sensitive to ambient or operating conditions
- Operate at a wide variety of internal temperature and pressure conditions

- Are scalable to a broad range of applications
- Are available for multiple types of catalysts

Fuel cells using these designs should penetrate many mass markets.

Drawings (see appendix B)

Figure 1 Diagrammatic representation of a side elevation of the improved anode with magnified views.

Figure 1A Rectangular flat plate porous flow through anode. The arrow represents flow of fuel/electrolyte mixture.

Figure 1B Magnified cross-section view of a diagrammatic representation of the gross microstructure of the porous flow through anode of fig 1A.

Figure 1C Magnified view of a diagrammatic representation of the fine microstucture of a porous substrate with catalytic coating of fig 1B.

Figure 1Ca Alternate microstructure of sintered particles. May be coated with, or made of, the catalyst.

Figure 1Cb Diagrammatic representation of a porous substrate with voids filled with catalyst supporting particulate surface area expanders. They may or may not be sintered in.

Figure 1Cc Diagrammatic representation of a porous substrate with voids filled with high surface area particles such as flakes. They may or may not be sintered in.

Figure 1Cd Diagrammatic representation of a porous substrate with voids filled with grown in nanostructures

Figure 1Ce Diagrammatic representation of a porous substrate with hollow walls and catalytic coating on just one side. The coating may be applied to both sides.

Figure 1Cf Diagrammatic representation of a porous substrate made of catalytic/ conductive material.

Figure 1Cg Diagrammatic representation of a porous substrate with nonconducting substrate with conductive/ catalytic coating.

Figure 1Ch Diagrammatic representation of a porous substrate with attached high surface area particles.

Figure 2a Side view diagrammatic representation of 2 sided anode with central less
dense, larger pore, high flow core for fluid ingress

Figure 2b Side view diagrammatic representation of a side view of a thick anode with
pore size gradient across the thickness for increased surface area without
increased resistance to flow

Figure 3a Side view anode with closely applied relative fuel barrier membrane at the
egress side

Figure 3b Side view anode with relative water barrier semipermeable membrane
applied to the ingress side

Figure 4a Diagrammatic representation of a fuel cell design with fuel separating
membrane in the pre-stack electrolyte circuit and a nonselective
permeable membrane between the anode and cathode.

Figure 4b Diagrammatic representation of the stack design of figure 4

Figure 5 The stack case showing one possible way of orienting the fluid and air
intakes and outputs.

Figure 6 Sketch of manifolds placement relative to the stack case.

Figure 7a Side view rarified core anode surrounded by two cathodes

Figure 7b Side view of a flow through anode surrounded by two cathodes

Figure 7c Side view of a single cathode paired with a more massive anode

Figure 8a Diagram of separated anodes and cathodes permutation demonstrating
differential number of anodes vs. cathodes and different flow speeds in
each compartment.

Figure 8b Diagram of separated anodes and cathodes permutation demonstrating
differential anode and cathode morphology for optimal performance.

Figure 9 The water, thermal, pressure management system

Figure 10 The assembled fuel cell system

Static Descriptions of Diagrams

Figure 1 the diagrammatic representation of the improved anode shows the anode in a flat
plate configuration as seen from the side, with magnified views. Wavy arrows (4) represent
the electrolyte and fuel mixtures flow through the porous body of the anode (2).

Figure 1B shows a magnified diagrammatic detail of a cross section view of the gross
microstructure of the porous flow through anode. (6) represents thin walls separating
microscopic flow through channels (8). The fluid electrolyte mixture (10) flows through the
space (8) by the wall (6) via pore(15).

Figure 1C shows a still greater magnification of the structure in 1b detailing the
composition of wall (6). In this particular example solid framework (12) is coated with a
catalytic/conductive thin film (14) creating complex(15). The fluid flows over the thin film
(14) through the channels (8).

Figure 1C(a) shows an alternate microstructure wherein micro particles (1) have been
sintered to form the body of the porous anode. The particles may have high surface are
morphology, the may be coated with a thin film catalyst. The coating may be

discontinuous. For a greater catalyst dispersion and greater ease of sintering the particles can be coated in a patchy or splattered/scattered pattern rather than continuously and/or the catalyst coated particles can be mixed with uncoated particles. In alternate embodiment, catalyst particles are mixed with particles with less or no catalytic activity.

Figure 1C(b) shows an alternative embodiment in which a porous framework (30) has voids filled with particles (32). These particles may be analogous to the current carbon supported platinum matrix or they maybe analogous to Figure 1C(a)'s catalytic coated sintered particles.

Figure 1C(c) shows an alternate embodiment where a porous framework (a) represented diagrammatically by (30) has the voids filled with high surface area flakes or particles (34). These flakes or particles may be composed of catalytic materials or alternately have a core of a different substrate than the catalytic coating.

Figure 1C(d) shows an embodiment in which porous substrate (30) has had nanostructures (28) such as nanohorns or nanotubes grown into space (8).

Figure 1C(e) is an alternate embodiment in which the porous substrate has hollow walls (16) with catalytic coating (14). The catalytic coating (14) may be on all sides of (16) including the interior (not shown). (18) represents a pore to the interior of the walls. (20) represents the interior of the walls.

Picture 1C(f) is a diagrammatic representation of a porous solid core substrate wherein the material (22) directly provides catalytic and conductive function without coatings.

Figure 1C(g) porous substrate (26) is composed of an inert material with catalytic and conductive coating (24) porous substrate.

In figure 1C(h) high surface area particles (27) are on the porous substrate(29)

In figure 2A a diagrammatic representation of a cross sectional view of a flat plate anode is shown. The anode is comprised of outer layers (38) with a very high surface area, small channel diameter and a large number of pores per inch for maximizing surface area.
This electrode embodies a central core (40) composed of similar material but with greater pore size diameter and relative lower density than in exterior (38) to allow for the influx of the fuel electrolyte mixture (36).

Figure 2A is a diagrammatic view of a cross section of a flat plate anode. In this case three distinct zones (42,44,46) are displayed. These layers demonstrate an anode design with the area facing the fluid ingress (42) being maximized for high flow low resistance conditions with large pores and less relative density. (44) is an intermediate zone with medium pores, medium surface area and medium resistance to flow. (46) is a high surface area, small channel relatively higher resistance to flow region maximized for surface area for catalytic surface area fluid interactions.

Figure 3A shows a cross section of electrode (48) with corresponding membrane (50). Membrane (50) is located in a downstream fashion from electrolyte (48) in the flow of the fuel electrolyte mixture (47). Membrane (50) has the properties of being semi permeable. It acts to selectively impede the fuel (51) while allowing the water and electrolyte mixture (49) to pass.

In figure 3B electrode (48) has a membrane (52) that is positioned upstream of the fuel electrolyte mixture flow path (47). Membrane (52) has the property of selectively impeding the passage of water(49) while allowing the fuel (51) to transmigrate into anode (48).

Figure 4 is a diagram representing a fuel cell electrode complex with electrolyte/fuel flow circuit. Showing an alternate embodiment in which membrane (56) is positioned in chamber (58) outside of the fuel cell stack. Fuel, water and electrolyte mixture enter by port (60). Membrane (56) selectively impedes water while allowing fuel to pass through it. The relatively fuel depleted egress (61) travels through conduit (64) into space (72). The space between cathode (68) and anode (76) is divided by porous non-selective permeable barrier (71). In this representation the side of cathode (68) facing away from the fluid has hydrophobic membrane (66) that allows airflow to permeate into cathode (68). Cathode channel (72) is a closed space, which experiences the relatively continuous influx of fluid (61). This creates hydrostatic pressure that causes a net egress of the fluid in space (61) into the space (74) across membrane (71). The relatively concentrated fuel mixture (63) is transported to anode space (74) via tube (62). There it permeates and filters through anode (76) to where it is collected in space (78) and returned via conduit (80) to fuel processing box (58). (54) represents the system by which additional fuel is added into stream (63) via injector (55).

Figure 4B is a diagrammatic representation of the association of stack components described in 4B individually. (82) is the manifold by which high concentrate fuel mixture is brought into space (96). Manifold (84) brings low fuel high water flow into space (92) by cathode (90), which is covered, on the contra lateral side with hydrophobic porous membrane (88) that allows air (102) to permeate into cathode (90). The fluid in cathode space (92) filters via hydrostatic pressure across permeable separation membrane (94) into space (96). The 2 fluids mix and permeate through anode (98) into space (100) where they are transported back to the balance of plant apparatus by manifold (86).

In figure 5 ones sees a corner elevation of the exterior of the case for the fuel cell stack (104). (104) has openings (106) along one face for the ingress of air with similar openings on the opposite face (110) for air egress. Case (104) on sides perpendicular to (106) has slots (108) for the ingress of the fuel electrolyte fluid and similar spaces along the opposite face (112) for the egress of fluids. In this diagram the electrode plates would be holding the interior of the case in water/airtight channels. The electrodes would be flat thin plates oriented parallel to face (104) between the various slots (106,108). An alternate embodiment the entering liquid and/or air can egress on the same side to form a counter current multiplier to preserve heat within the stack (not shown).

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In figure 6 the fuel cell stack case (104) is shown with one embodiments relative positions of air and fluid manifolds. Fan (103) blows air into manifold (128). The air passes through the slots in case (104) between the hydrophobic layers of the cathodes. The air egresses through manifold (124) over coil (126). The fluid ingresses through tube (116) into manifold (114) through slots in case (104) through the porous anodes out through slots on the contra lateral side of (104) into manifold (118) and out through tube (120).

Figure 7A through C show 3 alternate anode/cathode configurations/relationships.

In figure 7A anode (135) is comprised of high surface area external sections (138 and 142) with a relatively rarefied or even hollow area (140) is sandwiched between cathodes (136), which have hydrophobic membranes (132) on the contra lateral side. Depending on the direction of circulation, fluid enters or leaves through the space between the cathode (136) and anode (135). It filters through the porous walls of the anode (138 and 140) into or from the space (140). In this embodiment the fluid flow direction augments the mass transport of the hydroxyl species.

In figure 7B flow by anode (144) is sandwiched between cathode (136), which have hydrophobic membranes (132). Fluid flows into spaces (134) and by the anodes and cathodes.

In figure 7C relatively thicker anode (146) is paired with cathode (136). Fluid ingresses through anode (146) from the side contra lateral to cathode (136) and out through the space between anode (146) and cathode (136). It may flow in the opposite direction.

Figure 8A shows an alternate stack structure in which the anodes are grouped together in a chamber relatively separated from the cathodes. Injector (154) injects fuel into the fluid in space (152). This fluid is circulated into the anode room (153) where it flows into channels (156) through the anode (157) and into the spaces (158). The fluid then is collected into space (160). It is circulated into the cathode room (155) where it flows into channels (148) between cathodes (150). Air circulates between channels (151) on alternate sides of the cathodes from the fluid. Fluid then flows out into space (152). The significance of this diagram includes the fact that there are different number of anodes and cathodes. Flow channels (148) and (158) have differential flow characteristics including total cross sectional area and length designed to alter the fluid flow dynamics to maximize either anode or cathode function.

Figure 8B is a schematic diagram representing the flow circuit of alternate fuel cell stack embodiment wherein the anodes and the cathodes are grouped together relatively separated from one another. Pump (190) circulates fluid past (160), which injects additional fuel. The high fuel mixture flows through conduit (162) into the intra anode space (164). The anodes (168) are relatively short and thick compared to the cathodes (178). The fluid permeates and filters through anodes (168) into collecting systems (170 and 172). It flows by conduit (164) into the cathode chamber (175) and into the intra cathode space (176). Air flows through the contra lateral sides of the cathode from the fluid via channels (177). The fluid is collected in manifold (18) and returned to pump (190).

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Figure 9 shows a diagram representing a side view of the product water, and thermal management systems. Pump (204) circulates fuel-enriched fluid via conduit (206) into the fuel cell stack case (192). Fuel depleted, high water concentration fluid egresses from the stack (192) via conduit (196) into coil (198). (198) is an apparatus in which the fluid flows over a selective semi permeable membrane that allows the passage of water while retaining fuel and electrolyte dissolved components. As the fluid flows through (198) internal pressure causes a reverse osmosis like phenomenon, wherein product, water, is transpirated through the membrane of (198). The relatively water depleted mixture travels via conduit (202) back to pump (204). Fan (194) draws air through the fuel cell stack (192) between the cathodes. The air egresses over semi permeable membrane of apparatus (198) and out of the assembly at (200).

Figure 9A is a cross section of one embodiment configuration of the semi permeable membrane (198) wherein the membrane has been formed into tubes. The fluid flows through the lumen of the tube carrying water, electrolyte and fuel. The water under pressure transmigrates through the walls of the tube into air space (200) with the air blown from fan (194).

Figure 10 is a diagram representing the relative positions of the major fuel cell system components. Pump (120) circulates both air and the fuel/water/ electrolyte mixture. The air travels through conduit (222) to manifold (234). It then travels through the intra-cathode space inside of the fuel cell stack assembly (210). The air then blows over the semi permeable membrane of water and thermal management device (216). Pump (220) circulates fuel via conduit (224). This embodiment includes a pre cathode assembly (226). Additional fuel from tank (223) is introduced via injector (228) into fuel mixing chamber (230). The fuel then flows through fluid input manifold (208) into the intra anode space into the fuel cell stack assembly (210). It flows through the porous body of the anode and is collected in fluid egress manifold (212) where it passes through conduit (214) and through the water transpirating thermal and water management system (216).

Components and how they work.

Background: The current art anode inefficiencies:

In the most common forms of current art PEM and direct methanol fuel cells, the anode is composed of a mixture is finely divided, widely dispersed very small particles of platinum dust supported on, and adhered to, larger, but still microscopic, carbon particles. The substrate and catalyst complex is mixed with an adhesive binder to hold it together, fibers to increase structural integrity, and hydrophobic PTFE to help the egress of water. This mixture is applied to a structural support (in PEMs the support is often the electrolyte membrane itself).
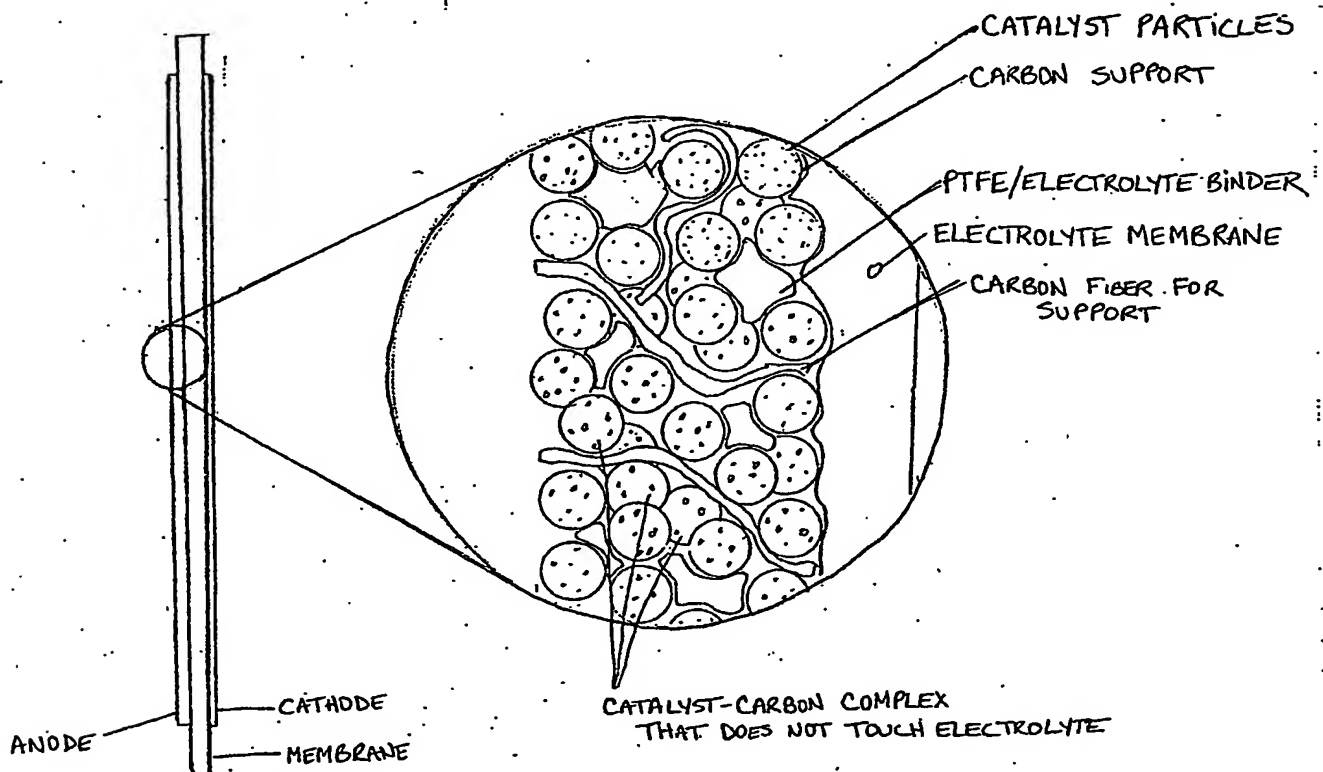

FIGURE A: SIDE AND EXPANDED VIEW OF CURRENT ART ELECTRODE AND ELECTROLYTE ASSEMBLY

Figure A.

This current design is functional and an improvement over earlier designs, but it is still vastly inefficient. This form of electrode suffers from several problems:

- Only a small fraction of the mass of the catalyst is available for catalytic activity. The catalyst molecules on the surface of the catalyst particle are the only part that touches the fuel. The rest of the mass of the catalyst is trapped in the interior of

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the particle where it cannot touch the reactants. We can calculate how much of the expensive catalyst is wasted in the above manner with some simple equations. Assuming the particles are roughly spherical:

The formula for the volume of a sphere is:
$$V= 4/3(pi)r3$$

The formula for the surface area of a sphere is:
$$SA=4(pi)r2$$

So, the ratio of surface area to volume of a sphere is:
$$S/V=3/r$$

Standard catalyst particle radii for PEM cells are 12.5-24.5 angstroms. Therefore, in PEM cells, the ratio of surface area to volume ranges from 1:4 to 1:8, with 1:6 as a median. **Only about 1/6th or 17% of the platinum is available as a catalyst, roughly 83% of the mass is wasted.**

The waste is much worse in direct methanol fuel cells. They require larger particles and heavier loadings. Direct Methanol cells generally employ particles with radii of generally 44-125 angstroms. Plugging these numbers into the above equations reveals that the surface are to volume ratios vary from 1:15 to 1:41 with a median of 1:28! **Therefore, only 3.6% of the catalyst is available surface area. The rest is potentially wasted by being locked inside the particle.**

· Further, part of the 3.6-17% of platinum that is the surface is additionally lost to the reaction because it is in intimate contact with inert substrate materials such as PTFE, glues, structural fibers, or the carbon support itself. This contact area excludes contact with the reactants. This further limits the usable, catalytic surface area. Losses from these contact areas can be conservatively estimated at about an additional 5%.

Additionally, another loss of catalytic utilization occurs because the reaction can only take place at the interface of the fuel, electrolyte, and catalyst. Only a fraction of the catalytic mass, left after the above reductions, encounters the electrolyte/fuel interface. Even in the case of the particles that do touch the interface of fuel and electrolyte, not all of the surface area experiences the interface. A significant amount of the catalyst does not even touch the electrolyte, and therefore does not participate in making electricity. Further, those particles that do experience a favorable interface produce or use water, thereby changing the fuel and water ratio in their immediate microenvironment, often decreasing catalytic efficiency.

● · The materials and mixture of the current art are fairly electrically resistant. This internal resistance substantially decreases electrical production efficiency and necessitates the use of conductive current collecting field flow plates that add sizeable cost and volume to the stack. A leading field flow plate technology also contributes considerable internal resistance.

- The carbon platinum mixture is a brittle composition of dust; a composite of powders. It is sensitive to vibration and mechanical, electrical and thermal stresses. Over time it tends to disintegrate limiting lifespan and efficiency.
- The fuel flows by or into the electrode, but not though it, so inert compounds can build up in the pores and physically block the fuel from reaching the catalyst further limiting efficiency.
- The individual parts of the fuel cell stack need a uniform, fairly exact degree of humidification in order to function. Because water is made at one electrode potentially flooding it, used up at the other drying it, and dragged by concentration gradients and electro-osmotic forces, extensive, costly and power robbing balance of plant apparatus has to be added. Even so, the simultaneous ideal humidification for each part is never quite uniformly achieved.

What is being patented: The improved anode design specifics

The basic structure will be a porous, electrically conductive electrode. A fluid can flow through the porous body of the anode, over a tremendous surface area. The anode should have good internal electrical conduction for integral current collection. The fuel mixture flows through the electrode, rather than just by or into it. In a preferred embodiment, the catalyst is a thin film coating on the extensive surface area throughout the sintered or metal foam porous electrode. (See Figure 1)

A variety of configurations and manufacturing processes can provide the needed porosity characteristics for flow through, high surface area design. Some permutations of the base microstructure configuration include, but are not limited to: open cellular, reticular, foamed, sintered, sponge, raney, nanostructure, vitreous, gel and/or aero-gel type substructures. The microstructure of the material is engineered with respect to porosity, pore size and shape, wall size, shape and thickness, as well as density to maximize surface area of available catalyst/substrate interaction, while preserving desired flow-through characteristics. An ideal configuration is an open cell thin walled metal sponge with a thin film coating. The important factor is that the pore volume be optimized for both flow and maximal electrolyte catalyst interaction. The walls may be hollow (figure 1Ce).

Materials for the substrate include porous conductive plastics, carbon compounds, ceramics, and metals. Stainless steel and nickel are to some extent preferred, but a variety of materials, mixtures and alloys are considered viable and even advantageous under specific conditions. These materials include, but are not limited to nickel, silver, molybdenum, titanium, ceramics, and carbon composites.

One of the most promising electrode manufacturing processes and the currently preferred embodiment uses a specific form of sintering. In this process, an inexpensive base material powder's particles are coated with a thin or discontinuous layer of catalyst. The catalyst coated base particles are then sintered into an extremely high specific surface area, flow through, coated porous electrode. A mesh of fine wire may be incorporated to promote conduction. This method bypasses the pore size limitations of electroless plating and creates very high surface area to volume ratios, but has the added challenges of maintaining the uniformity of the coating layer during the sintering process. Higher pressure, lower temperature techniques enable this method. There is considerable advantage from using odd shaped, high surface area particles such as twisted flakes.

A similar embodiment involves the use of a porous reticular metal substrate. Depending on the method of manufacturing employed to make the structure porous, additional means are usually employed to increase the available surface area. One such means includes sintering additional substrate(s) into the voids of the porous material (figure 1Cb). This is often necessary to achieve the needed surface area. This sintered material may be coated metal particles (figure 1Cc). Traditional carbon supported catalyst can be retained in the voids also.

In general, the goal is to produce an electrode that is less than 5mm thick and embodies 10 or more square meters per cc of catalytic surface area exposed to the fuel/electrolyte mixture. Average pore size goals are in the vicinity of .01mm, but there is a considerable range of possible sizes. Wall configuration is optimized for maximal thinness, preserving structural integrity. Depending on design and application specifics, there is considerable variation of practical densities, porosities, pore sizes, etc.

My current embodiments have also utilized a nickel or nickel chromium base reticulated foam substrate with a pore size of. 4mm. The available surface area is 5600cm2 per cc. Various methods for augmenting the surface area described herein are utilized to boost the available surface area above 10 m2/cm2. This is competitive with the some of the current art's calculated theoretic particle surface area. However, because the catalyst is used so much more effectively, much less catalyst may be used while achieving much greater efficiency.

The electrode may be directly composed of the conductive catalyst/ catalytic mixture (figure 1Cf) or may embody a different base material with a thin film coating(s) for conductive and catalytic function (figure 1Cg). The catalytic layer may be mixed with or layered on substances to increase conductive function. The porous substrate is either electrically conductive itself or it has conductive additives or coatings. It acts as a low resistance integral current collector.

Edge connection utilizes less material and simple off the shelf busbar design. It is the least expensive form of collecting system to manufacture. The walls may include a dielectric layer between two conductive layers to create a capacitor effect. The core may be a polymer/ceramic combined with a metallic conductor and/or catalyst.

Materials can be selected and blended/alloyed/ combined for conductivity (silver, molybdenum, etc), catalytic activity (platinum, palladium, ruthenium, manganese, silver, etc), cost (nickel, etc), corrosion resistance (stainless steel, etc).

The electrode may embody a porous substrate combined with pasted, grown, or packed material in the voids for the enhancement of the surface area. An example of this would be a metal sponge in which the voids contain the current art's carbon supported catalyst mixture. In another example, nanostructures such as nanotubes on nanohorns are grown onto the porous base metal into the voids for the dispersion of catalyst over a greater surface area (analogous to intestinal microvili) (figure 1Cd). The catalyst can coat or be integral to the nanostructures. In another example, a powder of high surface area low mass flakes or particles are pasted, sintered, etc directly into the voids or as the base structure itself (figure 1Cc).

The overall electrode can be manufactured in a variety of application-specific shapes and sizes. Categories of shape include, but are not limited to, flat plate, corrugated, tubular, conical, and cylindrical. The electrode may embody an area with less resistance to flow, surrounded by material optimized for surface area with more reduced flow-through

characteristics (figure 2a). This is one means of optimizing substrate/catalyst interaction while reducing the potential to form activity-depleted regions within the body of the electrode. Another such permutation involves manufacturing a pore size gradient into the electrode (figure 2b). The larger pores are on the fluid ingress side with the fluid flowing into progressively narrower channels. This design increases the available catalytic surface area while not significantly increasing resistance to flow and helps eliminate fuel-depleted regions in the thicker electrode designs. A preferred permutation of this would be to have the higher surface area side experience the fluid ingress. This way the highest fuel concentration is applied to the greatest catalytic surface area.

The electrode may include, be applied to, or function in conjunction with a membrane that relatively separates the fuel from the fuel/water/electrolyte mixture. There are several permutations of this design concept. If a membrane that selectively retards the passage of fuel is employed, it can be positioned on the fluid egress side of the anode (figure 3a). This will cause a relative increase in the fuel concentration of the mixture within the anode, increasing efficiency. This would also cause the fluid reaching the cathode to be further fuel-depleted thereby decreasing the potential for fuel crossover losses. The membrane, in this location, needs to allow the passage of hydroxyl ions. A membrane that allows the passage of fuel but relatively blocks water can be placed on the fluid ingress side of the anode to increase the fuel concentration within the body of the anode for the same effect (figure 3b).

There may be other catalysts specific to a separate, electrolyte replenishing reaction mixed in with, or in close association, with the electrode (described below).

Another embodiment employs the use of modified zeolytes as the anode body. These have ideal porosity surface area to volume ratios; they will need strategic modification to increase conductivity and to incorporate catalytic function. These methods include, but are not limited to, plating, deposition and mostly substitution of conductive and catalytic compounds. Zeolytes may be formed in the voids of a porous substrate.

A further permutation is the addition of a means to develop and control an electric and/or magnetic field that can increase mass transport, induce advantageous change in the electrode charge bilayer-.capacitor type. There may be an externally, or internally, applied means of creating an electric and/or magnetic field to elements of the stack such as electrically charged or magnetic compounds in or near the electrode. Such means include:
➢ A mechanism for inducing a cyclical fluctuation in the electricity exiting the stack. This could be as simple as a single rapid on/off chip in the electrical circuit. The off cycle would allow an increase in the local electric field potential, the on cycle would better allow the hydroxyl ingress. The ratio of time in each cycle can be set to optimize performance.
　　Similarly, a small charge can be introduced in either, or oscillating in both polarities to the circuit.
➢ An electric or magnetic field could be induced within the anode/ cathode space to hasten the mass transport of ions.
➢ A capacitor could be built into the body of the electrode for both of these functions, or it could be located adjacent to the electrodes. A permutation allows for an

intermediate dielectric layer within the layered porous substrate for the creation of a capacitor field.

How the anode achieves its results:
Efficiency Gains
Increased catalyst utilization efficiency- More available surface area per gram of catalyst

The catalyst, rather than being bound up inside particles and clogged with binders, etc, is a thin film coating on the extensive surface areas of the porous anode's channels (figure 1Ca). A much higher percentage of the catalyst can be presented as usable surface area, as compared to the current art. The fuel flowing through the microscopic pores in the electrode is presented with much more catalytically active surface area per gram of catalyst than is possible with current designs. Therefore much less catalyst is needed. In fact, because the catalyst is used with such higher efficiency, other catalysts besides platinum can be blended into the coating further decreasing platinum loading requirements.

Increased catalyst efficiency- Increased reactant/catalyst interaction

In all fuel cells, in order for the reaction to proceed, there needs to be a triple interface of catalyst, fuel and electrolyte. The reaction only occurs at the interface of all three compounds. If one of these components is not in direct contact with both of the others at the same time, that area will not contribute to the reaction. In the prior art, even under theoretical, (unachievable) ideal conditions, a very significant portion of the catalyst particles do not experience the triple interaction and are therefore wasted. They simply do not touch both the fuel and the electrolyte, as is required for the reaction (See picture A.)

These new designs apply the three components of the triple interface together universally. The fuel is dissolved into the electrolyte, so both are in continual and complete interface. Functionally, every molecule of fuel touches a molecule of electrolyte. The fuel/ electrolyte mixture then flows, through the porous electrode, over a tremendous surface area of active catalyst. The reactants are applied to the catalysts with orders of magnitude greater efficiency than in the incumbent technology they will replace. The area of triple interface is many times larger than in the prior art. Unlike the current art, 100% of the surface area is utilized 100% of the time. Simply put, unlike in the current art, much more of the catalyst, on a gram per gram basis, is actually used for making electricity. So, not only do these designs embody a much greater amount of useful catalytic surface area, essentially every molecule of that catalytic surface area experiences the triple interaction and contributes to the reaction. This is true even though less catalyst is used than in the current designs.

The increased catalyst surface area boosts the reaction rate, current density, and efficiency of the electrode. Therefore, it is possible for this design to use less catalyst to get more power, achieving and surpassing the goals set forth by the US Department of Energy for fuel cell catalyst loading.

Decreased concentration gradient losses

In the current designs, as the substrate is used and product water is made at the electrode, a relatively substrate depleted microenvironment forms around the catalyst. This decreases efficiency by limiting catalyst/substrate interaction. This is worsened because

relatively dead-end recesses, paired with the lack of flow through design of the current art, trap inert compounds and waste products. These substances can build up, keeping fuel from the catalyst. The 80% of air that is not oxygen, inert products in the fuel stream, and even the product water formed on the catalyst can get trapped and block fuel or ions from reaching the catalyst. Collectively this microclimate scale "gumming up" of the catalyst is termed 'concentration gradient loss.'

The flow-through design, surfactant nature of the electrolyte mixture, and oscillating fluid flow of the new designs all serve to break up concentration gradients before they can form, preserving maximal reaction rates.

Fluid flow is net forward but oscillates back and forth. This not only helps mix the fluids, it forces additional substrate to interact with the catalyst each pass. The net effect is that the substrate encounters much more catalyst area as it passes though the electrode, increasing efficiency again.

Reduced ohmic losses

More efficiency gains over the current art are gained in the area of electrical conduction. The current art, with its poorly conducting carbon and nonconducting PTFE components, suffers from a very high internal resistance. Simply, the base materials are relatively poor electrical conductors. This saps a significant percentage of the available voltage, wasting efficiency. To partially atone for this, designers of the present technology have had to include field flow plates that are also current collectors, adding cost and complexity while seriously dropping the power density.

Reduced ohmic losses are achieved by a number of means. First, in these new designs, the base materials and/or coatings are excellent conductors. Second, my design does not use binders on a discontinuous substrate, significant sources of electrical resistance in the current art. The internal resistance of my electrodes is essentially negligible. So much so that separate current collectors or field flow plates are not needed. They intrinsically have so little internal resistance that they can be edge connected. Incorporating various compounds directly onto or into the structure can further increase conductivity.

My designs also have much less electrical impedance. Impedance is the efficiency loss that occurs when electricity has to travel between the interfaces of materials with different conductivities. Impedance is minimal in my designs because the charge travels through only similar, intimately connected materials with very large interfaces. It can be further reduced, if desired, by including a small amount of the counterinterfacial compound to each layer. Additionally, my design has far fewer electrical junctures further decreasing impedance.

The dramatic increase in utilization efficiency and lowered catalyst requirements allow for the manufacture of fuel cells that **achieve and surpass the goals set forth by the US Department of Energy for fuel cell catalyst loading.**

Relative imperviousness to water balance issues

Current fuel cells are very susceptible to water balance problems. They need to maintain a very specific degree of humidification at each part. If any part gets too dry or too wet, it will malfunction. This problem is exacerbated because fuel cells operate in changing ambient humidity and encounter frequent temperature fluctuations. Furthermore, water is formed at one electrode, which can flood it, and water is used by, and dragged

from the other electrode, drying it. The membrane is subject to both problems. This creates a cell that seldom functions optimally, is quite sensitive to ambient and operating conditions, and requires additional, bulky and prohibitively expensive, water handling balance of plant equipment. The water handling equipment electrically parasitizes the fuel cell, dropping efficiency.

Fuel cells with my new design are not affected by the kinds of water balance problems that plague the current designs. The product water is washed away as it is made by the flow-through design. Because the water makes up a large percentage of the electrolyte, these designs are impervious to the degree of changes of water concentration that would paralyze the current art. Excess water is removed by transpiration and/or evaporation elsewhere in the system with a self-regulating system that does not draw power. There is no need for the power drain of parasitic water balancing equipment required by the current art. The natural synergies of this design obviate the need for most water handling equipment, further substantially decreasing equipment requirements and cost while boosting efficiency.

Crossover dangers and efficiency losses

A problem with most fuel cell designs is that some of the fuel can get by the anode unused and wasted. This is called crossover and is deleterious in two fashions. First, the unused fuel represents wasted efficiency and increased pollution. Besides being wasted by not getting used, some of the crossover fuel can oxidize at the cathode further dropping efficiency.

My designs reduce fuel crossover efficiency losses by a series of features. In embodiments with anode to cathode flow direction, a large percentage of the fuel is used in the anode in a 'first pass effect.' Fuel utilization is augmented by specific flow rates and the oscillating flow. The fuel mixture is brought back and forth across the anode, utilizing a greater percentage of the available fuel, increasing the first pass efficiency. The little fuel that does make it through the anode becomes substantially diluted by the product water made at the anode. In the case of methanol, for every molecule used, 5 water molecules are made. Any fuel that makes it through the anode becomes diluted tremendously. For these two reasons, the cathode only experiences a relatively fuel depleted environment. Additionally, the cathode has catalysts with selectivity against the anode reaction, reducing crossover efficiency loss. The cathode also may include a number of fuel excluding technologies described in a separate application.

The second crossover problem is dangerous mixing. When crossover occurs, the current fuel cells can mix explosive fuel with oxygen in an electrically charged and heated environment. This dangerous condition plagues all low temperature fuel cells.

PEM design uses a tremendous area of membrane to separate the hydrogen from the oxygen. The membranes are paper thin and semipermeable. The gasses are hot, pressurized, and create caustic, reactive intermediary compounds that permeate the apparatus. Over time, the membranes develop leaks that allow mixing of hot pressurized hydrogen and oxygen inside a hot, electrically active device. It is a potential recipe for an explosion. Even if all other cost and efficiency problems were solved, there is still this dangerous problem with which to contend.

Fuel crossover mixing only occurs in an almost imperceptible degree in my design. The slight extent that it can happen is not unsafe, deleterious, or even significant. Such a small amount of methanol evaporates from the cathode that the fuel is diluted into literally millions of times more air volume so that there is no combustion risk. Further, my designs employ multiple means of reducing the amount of fuel that gets to and into the cathode.

In a preferred embodiment described below, before the fuel mixture enters the fuel cell stack, it goes though a series of separators that serve to channel concentrated methanol to the anode, away from the cathode (Figure 4.) The fuel injectors also feed into only the anode stream. The product water further dilutes any fuel left in the cathode stream. The cathode experiences a fuel-depleted environment, so very little of the fuel is available for crossover. The potential for fuel crossover is still further limited by the fuel-phobic nature of the materials within the body of the cathode. Very little fuel reaches the cathode, even less penetrates it, and the little that gets through is immediately washed away by such a hugely greater volume of air that it stays orders of magnitude more diluted than its combustible concentration.

Moreover, because the fuel is in liquid form, it does not mix in the same hazardous way that gasses do.

Decreased mass transport losses

In the current art, a significant bottleneck for the electricity producing reaction is the time it takes for the ions to cross the electrolyte. This is termed 'mass transit losses.' These new designs enable and employ a variety of ways of reducing this cause of efficiency loss. The oscillating flow mixes the electrolyte promoting faster ionic diffusion. The buffers and dibasic compounds can release extra ions as needed at the appropriate electrode should the local microclimate become depleted. Also, the electrolyte replenishing means keeps the ionic concentration optimal. These electrodes allow for the use of a means for inducing an electric field across the electrolyte to hasten transport (below.)

Electric field modifications

Within the anode, a negative charge builds up. Because like charges repel, this negative charge concentrates at the surface of the electrode. The surface charge concentration creates an electric field that attracts positively charged elements to surround the metal. In my fuel cell, this charge bilayer serves to augment fuel catalyst interaction by attracting negatively charged intermediary compounds to the catalyst. It can also be disadvantageous in that it can repel the needed hydroxyl ions.

These fuel cell designs enable the incorporation of a variety of means for manipulating the charge bilayer to favor increased fuel molecular /catalyst interaction and/or increased ionic diffusion rates.

In systems where additional efficiency gains are desired, the electrolyte in the anode-cathode space can be subjected to an electric/ magnetic field. A capacitor can be situated next to, or within, the electrodes to hasten the diffusion of the negatively charged hydroxyls toward the anode (improving an otherwise rate limiting step) and limit the fuel and intermediary compounds interaction with the cathode. The capacitor also can provide the electricity for the refresh cycle and help smooth out natural current fluctuations.

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Other features that are being patented
The refresh cycle
<u>Background</u>

Catalysts suffer from poisoning by common contaminants found in many fuel stocks. They are even poisoned by their own reaction intermediary compounds. Poisons include a variety of sulfur and carbon based compounds. Of special importance is carbon monoxide as it is a common intermediary compound of carbon-based fuels such as methanol. Overtime, these substances adhere to the catalytic particles of the current art electrode, degrading their performance and limiting the lifespan. **Sensitivity to poisoning seriously limits the feasibility and commercial viability of the current art.** It is especially a problem in fuel cells that use currently available fossil fuels and natural gas derivatives. They have relatively high amounts of sulfur compounds and complex hydrocarbons that form a variety of toxic intermediary compounds. To partially atone for this problem, manufactures are forced to incorporate expensive, additional balance of plant apparatus such as fuel stock scrubbers, reformers, shift reactors, and advanced filters. These are bulky and expensive.

They escalate cost, inefficiency, maintenance requirements, and pollution. The extra machinery also requires energy to run that is parasitically drawn from the fuel cell's output. The expense of the additional equipment obviates the advantages of being able to use readily available fuel stocks from the current infrastructure.

In addition to incorporating compounds into my electrolyte and catalytic coating to help solve this problem, my fuel cell design is uniquely suited to incorporate a refresh cycle that cleans the catalysts of poisons. In the refresh cycle, a specific electric charge is forced back through the electrode. At the catalyst, a specific amount of oxygen and/or hydrogen is formed, depending on the polarity of the charge. Especially the oxygen physically and chemically frees the catalyst of adherent toxins refreshing its function. The gas then dissolves or bubbles into the electrolyte and is cleared in the transpiration or venting equipment. It can also be used by the fuel cell.

The hydrogen can be caused to from at a specific, separate, electrode incorporated for the refresh function for appropriate handling. Studies are currently underway to evaluate hydrogen as a cleaning agent also. The electricity may be fed to the electrode with positive, negative, or alternating polarity for maximal cleaning/refreshing power with minimal gas production.

In the currently preferred embodiment, just enough gas is evolved to totally coat the catalyst in minute bubbles. The bubbles are small enough that buoyancy does not overcome the surface tension holding them to the catalyst. At the end of the refresh cycle, the small quantity of gas may dissolve, bubble off, or be reabsorbed at the electrode utilizing the fuel cell's own oxidation-reduction reaction.

In another permutation, enough gas is evolved to purposely cause bubbling of the gas off the electrode.

An interesting permutation utilizes the fuel cell's electrodes to create the hydrogen and oxygen in a new form of internal reforming. Using specific borohydrate fuel, hydrogen is made at the anode and oxygen is made at the cathode. Electricity is used to crack the hydrogen and oxygen out of the fuel stock. Then the hydrogen is consumed

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to produce water and heat. The net energy output is derived from the difference in enthalpies from cracking the fuel versus that for the hydrogen to water reaction.

There is added advantage when the refresh cycle is incorporated into the operation cycle. That is, there are extra electrodes in the stack that are rotated through an off/refresh cycle. For example, the stack calls for 100 MEA equivalents but incorporates 103. Two of the extra produce the power to clean and refresh the third. A microprocessor rotates the roles of the electrodes as the fuel cell operates. This allows the refresh cycle to occur at higher temperatures and prevents build up, ensuring the greatest operation efficiency, while decreasing down-cycle maintenance time.

An alternate method is to have the refresh cycle occur as part of the start up and /or shut down procedure or as part of scheduled off cycle maintenance.

The cell can be caused to run hotter for refresh cycles by limiting the cooling flow or amount of cooling coils in the flow circuit.

Unlike the current art, this stack design is also amenable to periodically being flushed with specific gasses and/or liquids cleaners to refresh the catalysts.

The electrolyte

The fuel/electrolyte mixture may take advantage of a variety of novel improvements not employed by the current art. The first is the incorporation of solvents and/or detergents that help clean the electrodes of poison and microclimate concentration gradients. The additives will help dissolve compounds that could otherwise precipitate out and cause a filter cake that can physically clog the filters or electrodes. They can break up the surface tension of product water globules, reducing efficiency sapping microclimate concentration gradients. In the current art direct methanol cells, without flow through design and cleaning additives, it is these gradients that limit designers to larger, more wasteful platinum particles.

The electrolyte may be oxygenated to further enhance cleaning.

The electrolyte of current design cells relies on the diffusion of ions through a medium to complete the separated redox reaction that creates the electric charge potential. The medium has to restrict the passage of parasitic internal electric currents. The mass transport of ions through the electrolyte is one limiting factor on overall reaction rates. Because the overall ionic charge balances must remain neutral, an ion cannot react at one electrode until one is made at the other.

In this improved electrolyte, buffers, hydroxyl carriers, and multibasic (polyhydroxyl alkalis) compounds are incorporated into the fluid. These increase the mass transport efficiency. They allow a degree of charge-balanced hydroxyl supply margin that can hasten focal reactions restricted by the above dynamic. When focal conditions cause hydroxyl-depleted regions additional hydroxyls can be locally drawn from the electrolyte chemical reserve.

In the incumbent technology, the flow of charged ions through the electrolyte, and of electrons through the wire, create a circular path. The reaction rate of any part of the overall process is restricted to the rate of the slowest step of the process, which is the passage of ions through the electrolyte. In the new method, extra hydroxyls are available from the electrolyte to promote a faster basal rate at the rate limiting focal regions.

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Sections of the electrode experiencing a relative dearth of hydroxyls can 'pull' them from the chemical reserves of the buffers, polyhydroxyl alkalis, and hydroxyl carriers in the electrolyte. Temporary fluctuations in microconditions will no longer impact the overall rate. A degree of elasticity is introduced into the system.

With the advantages of overall increase in hydroxyl transport by polyhydroxyl alkalis, buffers, and hydroxyl carriers, it may be possible to reduce the limitation of charge mass transport. This would speed the overall reaction and possibly allow the anodes and cathodes to be spaced further apart to reduce crossover electrical currents.

Electrolyte refreshment

In the current art circulating electrolyte alkaline fuel cells, the electrolyte slowly reacts with product and ambient CO_2 to form an inert product. The most common example of this reaction is:

$$KOH + CO_2 \rightarrow K_2CO_3 + H_2O + heat$$

This process impedes the fuel cell function in a number of ways:

- ➤ The concentration of KOH goes down reducing its efficiency as an electrolyte.
- ➤ The dissolved K_2CO_3 further dilutes the electrolyte.
- ➤ K_2CO_3 precipitates out potentially gumming up the electrodes.

In order to extend the maintenance free intervals, current art alkaline fuel cells depend on additional balance of plant apparatus to 'scrub' the CO_2, to filter out the K_2CO_3, and to replenish the electrolyte.

My design takes advantage of numerous intrinsic synergies to reduce the rate of electrolyte degradation. The first way this fuel cell can avoid this problem is to not use carbon-based fuels. My design is flexible with respect to the range of possible fuels. Here are many fuels that are not carbon based and therefore do not make CO_2. Hydrogen, $NABH_4$ are examples of fuels that don't create CO or CO_2 and therefore will not degrade the electrolyte. Methanol, however, is a carbon-based fuel with a low, but inevitable, carbon dioxide production ratio.

The process of electrolyte degradation is a reversible reaction that tends toward equilibrium. As the electrolyte goes through its flow cycle, it experiences a surprising variation in local conditions. Upstream from the anode, it is relatively concentrated, cool and has a high fuel load. As it passes through the anode, it is heated, the fuel is depleted, and for every molecule of methanol used, five water molecules dilute it. This drastically alters the concentration, PH and temperature of the fluid. At the cathode, there is a relative increase of hydroxyls and the water is used up increasing pH and concentration. The electrolyte is also subject to cooling from the mass flow of air at the cathode. In the water-handling chamber, the mixture is further cooled and concentrated. Then more fuel is added.

The fluid is subject to a variety of widely changing local conditions. Some of these conditions favor, or come closer to favoring, the reactant side of the above equilibrium. One specific location that relatively favors the renewal direction of the reaction equilibrium is just downstream from the anode. Here, there is a relative excess of heat and water and relatively low pH and KOH concentrations. At this point, a specific catalyst can be

incorporated. Because conditions favor the return reaction a catalyst here would renew the electrolyte. This catalyst can be included in or near the surface of the egress side of the anode. Solubilizing agents can be added to further tip the equilibrium favorably.

If local conditions do not achieve the ability to favor the renewing direction of the equilibrium, a fraction or the entire electrolyte can be caused to flow through a side path with a countercurrent multiplier, to augment favorable conditions.

Stack design

The fuel cell stack is housed in an inexpensive molded plastic box-like case. Construction and materials are quite similar to those for the casing of lead acid batteries. The anodes and cathodes are edge sealed into channels in the interior walls of the case. The case has slots on opposite faces for the ingress and egress of liquid and air. In the currently preferred embodiment, the air ingress and egress slots are on opposite sides of the case, perpendicular to the liquid's slots. (figure 5) Simple manifolds carry the substrate fluids to and from the appropriate slots. (figure 6) The manifolds are segmented to provide long thin channels that prevent internal electrical currents. In an alternate design, liquid ingress occurs next to egress to provide an intrinsic countercurrent heat exchanger.

In current low temperature designs, there are equal numbers of anodes and cathodes. A source of current art fuel cell efficiency loss is anode vs. cathode kinetic mismatch. In many systems, the anode could function at a much higher rate, but the slower cathode limits it. In several instances, the differential in electrode kinetics is two orders of magnitude. Designers adjust for this by differential catalyst loadings on each electrode.

In the current art, the electrodes are grouped in functional units of single anode/cathode pairs. The functional pairs may be arranged in two fashions. They may be placed so that the anodes and cathodes alternate in an every-other pattern, with like electrodes facing one another across the field flow plate.

In my design, the electrodes can be arranged in novel, ways for additional benefit. Differential kinetics can be partially or whole balanced, not just within the range achieved by varying the loadings, but also by the use of additional cathodes or more massive cathodes (picture 7a-c.) The anode may be surrounded by cathodes that balance the reaction kinetics. In one permutation the anodes and cathodes are relatively separated. (picture 8.) In this permutation, the disadvantage of vastly greater ion transport distance is surmounted by a variety of considerations:

> Increased flow rates
> Eliminated electrical crossover
> Decreased fuel crossover
> Increased fuel utilization at the anode
> Ability to maximize each type of electrode's performance without the need to account for the other.

The anodes and flow can be shaped for maximal efficiency without having to mach the anodes size or shape and vice-versa.

In the illustrated examples (figure 8a+b), the flow chamber for the cathodes has a greater cross-sectional volume than that for the anodes. This slows the fluid flow down to compensate for slower reaction rates at the cathodes, balancing the system. Because the hydroxyls are delivered by flow rather than by diffusion, the anodes can be designed

thicker, allowing for wider pores to decrease resistance to flow and increased total surface area for greater fuel utilization. A side circuit can be added to recirculate unused fuel leaving the anode.

Electrolyte circulation route

For added benefit, my design can embody an electrolyte flow path that hastens the mass transport of hydroxyls speeding the overall reaction. Unlike the prior art, my fuel cell can have net current flow within the stack travel generally from the cathode to the anode. This helps cool the anode and, more importantly, speeds the rate at which the ionic transport occurs.

One permutation of the flow design is to modify the pump to create an oscillating flow. The fluid flows back and forth in small amplitudes, but generally forward. This has many advantages. It further breaks up concentration gradients, mixes fuel, cleans out precipitants, and hastens mixing. Fuel flowing through the anode is dragged back and forth over the catalyst, increasing utilization. Mass transport is hastened by three means with this flow pattern. The net flow washes the ions in the right direction, the mixing agitation increases diffusion rates, and the backward flow temporarily increases the concentration of ions, increasing diffusion pressure. The oscillating flow and flow-through function not only increase fuel/catalyst interaction for better fuel utilization, they wash away the product water, and increase the rate of ionic diffusion. They also can reduce precipitate build up.

In another embodiment, the fluid circulates in an anode to cathode direction. The anode uses the fuel and adds water to the mixture, so the cathode experiences only a diluted, fuel-depleted environment that is highest in the water that it needs. This promotes cathode efficiency while decreasing fuel crossover losses.

In a currently preferred embodiment (figure 4), the fuel /electrolyte/ water mixture passes through a separating chamber. Here, it is separated into a fuel rich (A) stream and a water rich (B) stream. The A and B streams pass into the anode-cathode space on either side of a porous flow limiting permeable barrier. This barrier is just obstructive enough to keep the A stream from appreciatively mixing back into the B stream. The B stream comes in on the cathode side of the permeable membrane, supplying a low fuel, high water environment that is ideal for cathode function. The cathode uses the water to make hydroxyls for the electrolyte. This is a closed space, so the only egress is through the permeable barrier into the anode side. The now concentrated and hydroxyl rich B stream flows into the anode side, where the anode is receiving fuel enriched fluid ideal for its function. The mixed fluid egresses through the porous anode bringing high concentration of fuel and hydroxyls to optimize its function. The cathode does not experience much fuel, limiting crossover. The net flow is in the direction of the hydroxyl diffusion, speeding up mass transport.

Balance of plant apparatus

The balance of plant apparatus is the ancillary equipment that is necessary to support operation of a fuel cell and condition its outputs to usable forms. Balance of plant apparatus includes controllers, heat exchangers, fuel reformers, shift reactors, humidifiers, dehumidifiers, pumps compressors, regulators, power conditioners, tanks, valves, pipes, hoses, sensors, thermal regulators, manifolds, etc.

26

In the current art, the massive balance of plant apparatus dwarfs the actual fuel cell stack. For example, a fuel cell stack the size of a toaster may require balance of plant apparatus the size of a clothes washer. **Much of the prohibitive cost of fuel cell production is for the balance of plant, not for the fuel cell stack!**

My fuel cell does away with the need for 70-90% of the balance of plant apparatus. This alone may more than halve the cost and maintenance, while boosting efficiency and power density.

The water handler

All fuel cells make product water and heat. Current PEMs and direct methanol fuel cells, being very sensitive to water balance issues, require substantial parasitic water handling balance of plant equipment. They also employ balance of plant apparatus for preheating the reactants and reformers, as well as for removing excess heat. The cost, power drain and maintenance requirements of these systems are a viability barrier to most applications of these designs.

This new fuel cell is uniquely suited to a novel, inexpensive, simple, non-parasitic means of handling and removing product water. First, unlike the current art, the flow through anode operates efficiently at a vastly wider range of incoming water concentrations. It does not require stringent control over a changing narrow range of humidification. Unlike the current art, the component efficiency is not degraded by the expected concentration microgradients and fluctuations intrinsic to normal operation. It is unaffected by even the range of fluctuations that can paralyze conventional models. Second, the flow through process washes away excess water build up especially with additional benefits of detergents/surfactants and mixing of the oscillating flow. There is no need for the current art's equipment that collects exiting water to humidify the input, or for all the equipment that regulates this process. In my design, the product water causes its own regulation.

The electrolyte circulation passes, in whole or in part, over a semipermeable membrane. This membrane has similar functional characteristics as reverse osmosis water purifying filters. When subject to an aqueous fluid under pressure, a percentage of the water is selectively passed through the membrane. The dissolved and ionic compounds are retained. In the preferred embodiment, the membrane is formed into a coil of a tubular element. (See figure 9). The electrolyte flow circuit is a closed loop. As fuel is broken down into a net excess of water, the fluid volume in the closed loop rises. This increases the pressure. The water handing membrane is capable of reverse osmosis at the higher pressures exuding, filtering, and transpiring the excess water. This also controls the pressure build up.

The membrane composition is selected based on the application parameters and fuel type. Generally it employs layers with hydrophilic and hydrophobic character. It can be composed of a varied substances and work by a variety of means, including pervaporation. Activated nylons, cellulose acetate, nitrocellulose, oleophobic preparations, nafion™, polyvinyl fluorides all have usable characteristics. The preferred embodiment uses a variant of off-the-shelf water purifying reverse osmosis technology with additional hydrophilic layers.

Thermal management

The stack is first cooled by the air blown over the large surface area of the cathodes. The second cooling step occurs because the water management coil is in the path of the airflow that exits the cathodes. The transmigrated water on the outside of the tubes is subject to airflow. The air blows over the tubes moistened by the transudated water, causing the water to evaporate- a very effective and self-regulating cooling means. The flow of air serves to carry away the product water and to cool the fluid by both evaporation and conduction. Water build up, thermal management, pressure control, and electrolyte concentration are all managed by this same simple, compact, elegant system. It is self-regulating and works without parasitic current drains.

As the fuel cell experiences a heavier electrical demand, water production is accelerated, raising the pressure and temperature, hastening the reverse osmosis water removal and cooling function automatically, synergistically. All without the need for controls, parasitic energy input, or even many materials. In essence, all that is needed to handle all those functions is a well-designed coil of modified off-the-shelf materials. Unlike current art, these functions happen automatically with much less equipment and no energy drain.

This fuel cell requires much less balance of plant apparatus. This substantially reduces costs, size, parasitic energy drains, and maintenance requirements, while boosting efficiency, power density, and lifespan.

Miscellaneous balance of plant

A problem with other low temperature fuel cell designs is the dependence on pressurized gaseous fuel stock. Gasses are hard to work with, necessitating the expense of complex, expensive, less reliable, and less safe machinery. These electrodes can operate in gaseous systems, but work best with liquid substrates. There is no need for reformers, shift reactors, fuel processors, high-pressure tanks, field flow/bipolar plates, preheaters etc.

Again, the bottom line is that fuel cell systems with these designs will be smaller and less expensive, they are also more powerful, reliable, and efficient than the current art.

Appendix A

Problems specific to PEM type fuel cells

Hydrogen fueled PEM technology currently leads the industry. It has many important advantages, but is not without multiple weaknesses. The following is an in depth discussion of the challenges facing PEM technology.

The first problem with PEMs is, as above, they are too expensive from both a materials and manufacturing perspective. PEMs require platinum. Platinum is very rare and its supply is mainly restricted to the former USSR and South Africa. If we start mass-producing platinum containing devices, the supply/demand curve will shift, causing the already prohibitive price to rise. Additionally, platinum mining is very damaging to the environment. Ten tons of earth are moved for each ounce of recovered platinum. Platinum, as a catalyst, is also sensitive to poisoning compounds commonly found in most fuel stock.

To lower platinum requirements, PEMs use esoteric components such as nanoparticles. Again, this is expensive and not easily lent to mass production. Further, these advanced composites are sensitive to damage from the vibration that can be expected from some applications. Also, the electrodes have a high internal resistance that decreases efficiency. This necessitates the use of expensive, inefficient, and bulky field flow plates. In fact, a majority of stack volume and cost stems from field flow plates.

The membranous electrolytes are expensive and too sensitive to water balance and temperature issues.

PEMs are very susceptible to water balance problems and need to have a specific degree of water concentration at each part. If the parts get too dry or too wet they will not function. This problem is worsened because they operate in changing ambient humidity and encounter frequent temperature and load fluctuations. Furthermore, waste product water is formed at the cathode, which can flood it, and water is dragged from the anode drying it. This creates a cell that is essentially never functioning optimally, is quite sensitive to ambient and operating conditions, and requires additional water handling equipment that parasitizes the system's electricity. The water balance issues decrease efficiency in three ways. First uniformly optimal humidification is impossible to maintain, so the cell does not run at its best. Second, there is the additional manufacturing and maintenance costs of the necessary additional water balance equipment. Third is the parasitic energy loss from running that additional equipment.

Aside from water balance issues, the stack needs to maintain a delicately balanced interface of solid, liquid, and gas components, called the triple interface. Varying temperature, pressure, humidity, and shifting load conditions change this balance. Any perturbation of conditions slows or stops the energy production.

The PEM design suffers from many sources of efficiency loss. These include activation energy, internal resistance, and concentration losses. Resistance to the flow of ions in the electrolyte and resistance to the flow of electrons through the electrode materials create an internal resistance that drops actual voltage. Concentration loss occurs on a microscopic surface level. As the electrode consumes the reactant, product water is produced on the catalyst. The concentration of reactants near the catalyst becomes diluted by the product water, causing a loss of voltage potential. The current arts non flow-through electrode exacerbates this problem. The non-fuel substances become trapped in relatively

blind channels, excluding new reactants from penetrating the electrode to create more electricity.

Another important obstacle to widespread PEM commercialization is hydrogen. PEMs are not fuel flexible; they can only run on hydrogen. As dicussed above, hydrogen is volatile, explosive, and expensive to store. It makes metals brittle and escapes trough microscopic cracks. Storage techniques are too bulky, expensive and/or risky. Further, hydrogen is a very non-dense gas. To get a useful amount of hydrogen in one place it has to be compacted under tremendous pressure, which increases the types and likelihood of potential mishaps. Cryogenically cooling it into a liquid uses up much of the energy stored in the hydrogen. The refrigeration has to be maintained; otherwise the liquid hydrogen evaporates in its closed tank creating tremendous pressure. Moreover, there is no adequate infrastructure for hydrogen storage and distribution.

The final barrier to PEM cells is also the least acknowledged. PEM design uses a membrane to separate the hydrogen from the oxygen. The membranes are paper thin and semipermeable. The gasses are hot, pressurized, and create caustic, reactive intermediary compounds that permeate the apparatus. Over time the membranes develop leaks that allow mixing of hot pressurized hydrogen and oxygen inside an electrically active device that makes sparks. It is potentially hazardous.

Even if all other cost and efficiency problems were solved, there are still significant challenges to address. PEM technology has enough advantages that it will capture a significant market share, but other technologies have equal or greater potential.

Appendix B

FIGURE 1: DIAGRAMMATIC REPRESENTATION OF THE IMPROVED ANODE WITH MAGNIFIED VIEWS.

12
14
15
8

Figure 1C

Detail of fine microstructure

9
8
10
15
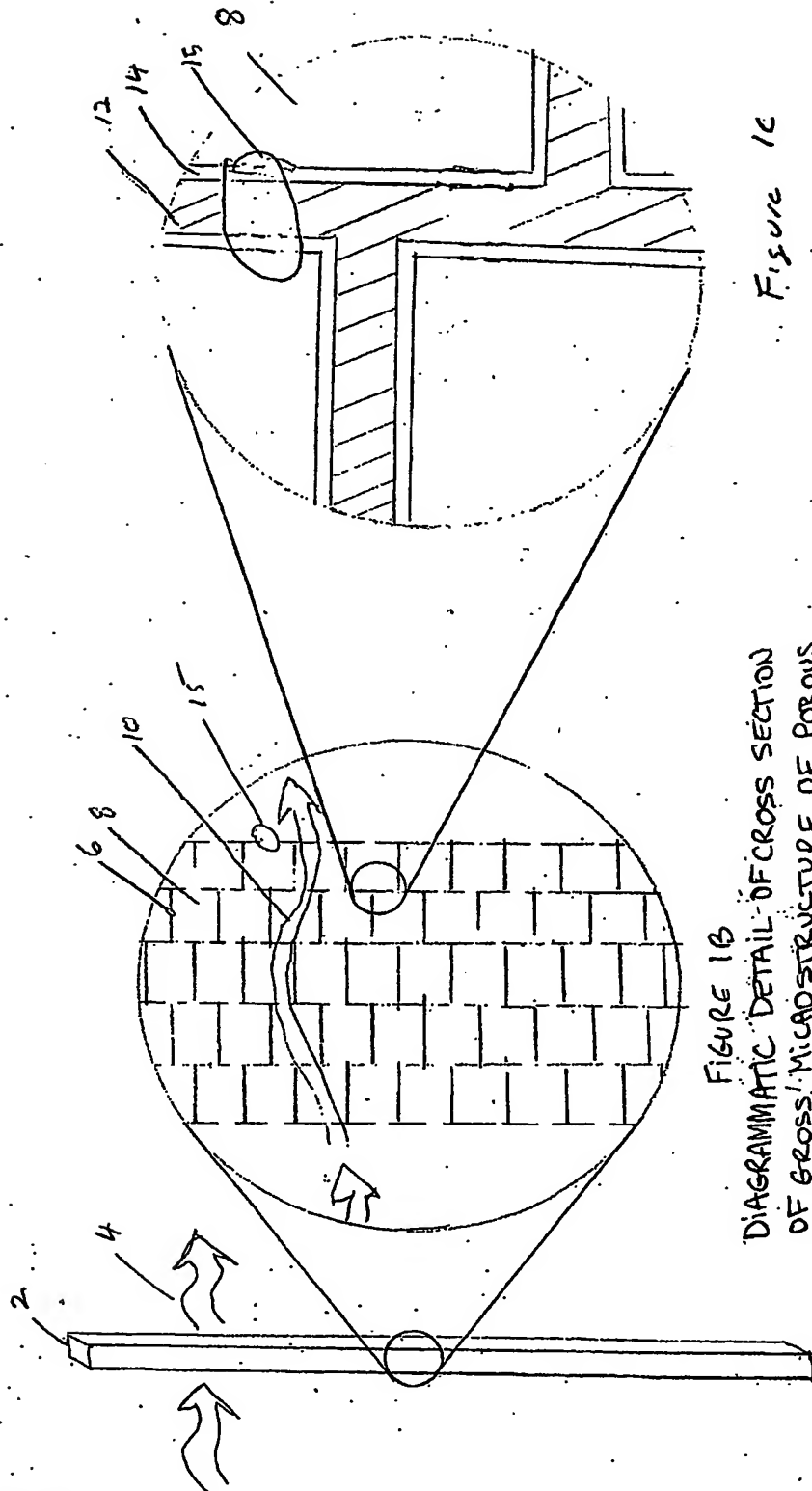
FIGURE 1B

DIAGRAMMATIC DETAIL OF CROSS SECTION
OF GROSS MICROSTRUCTURE OF POROUS.
FLOW THROUGH ANODE.

4

2

FIGURE 1A

Anode Flat plate
Side view
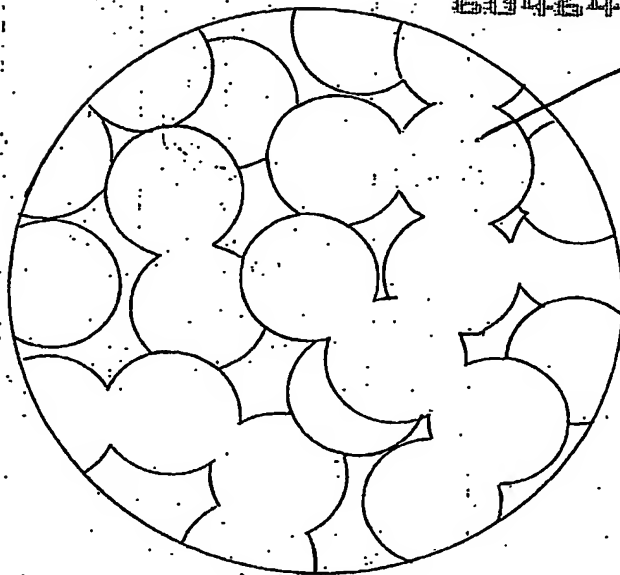
1C (a)
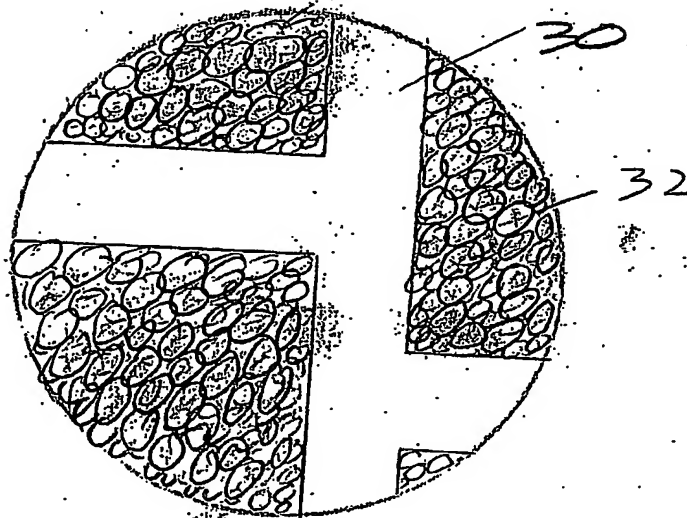
1C (b)
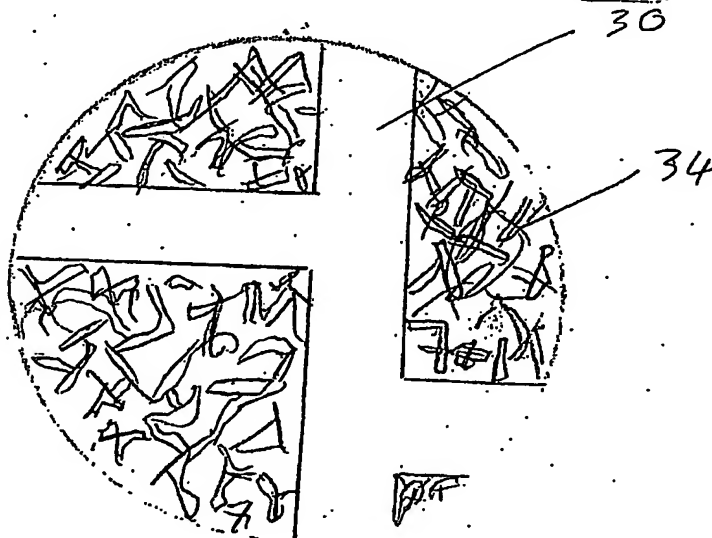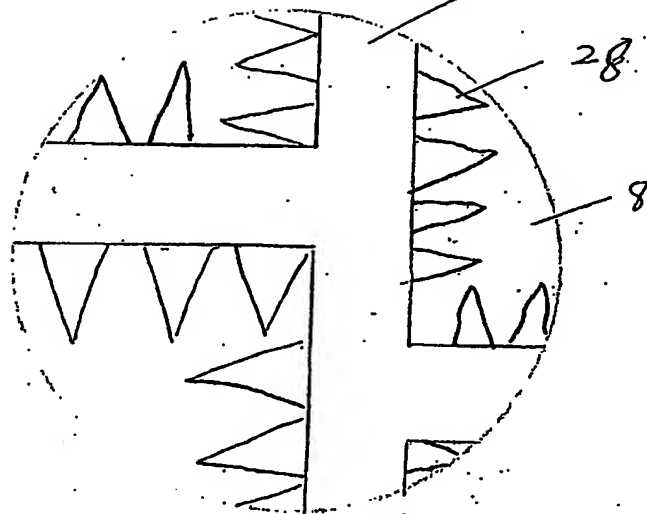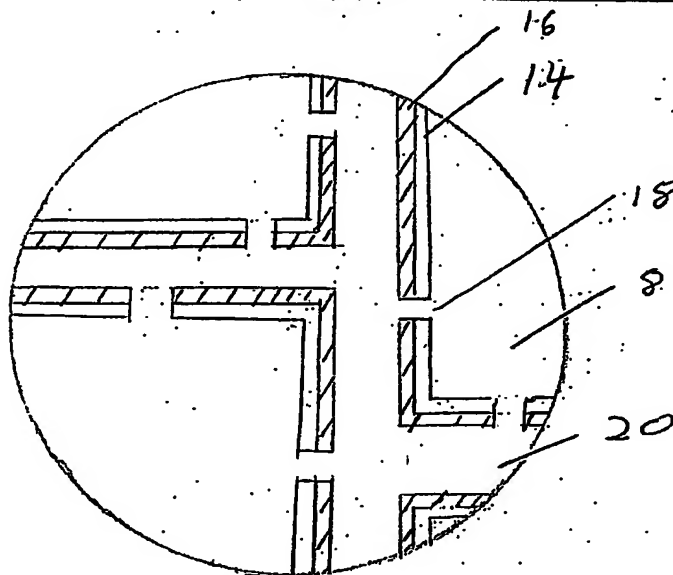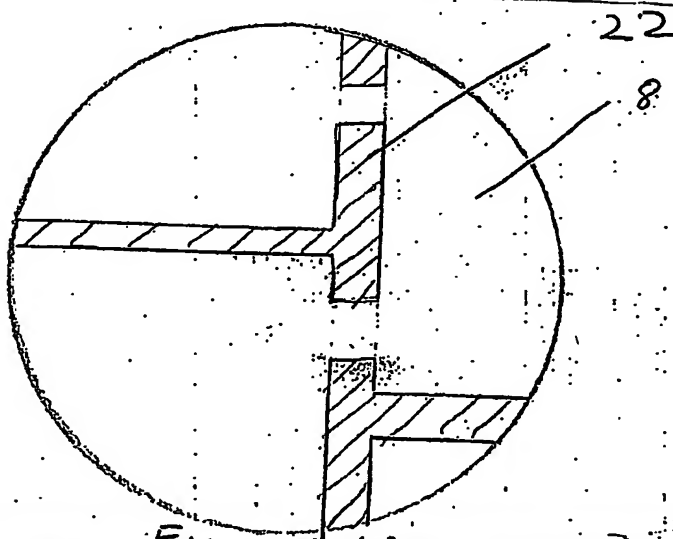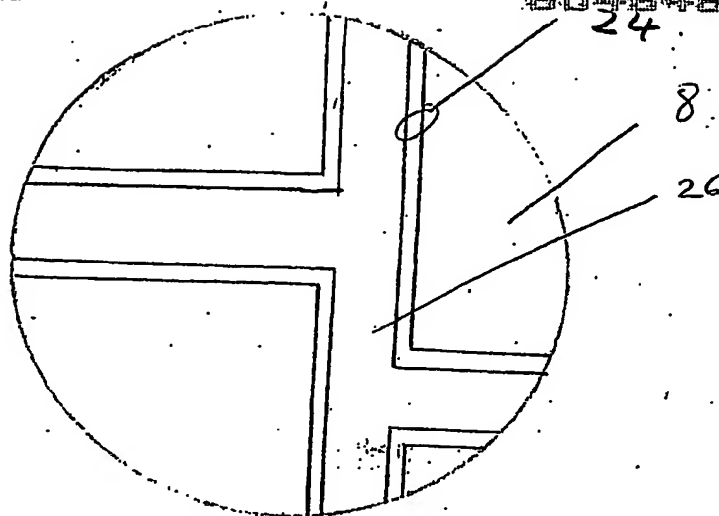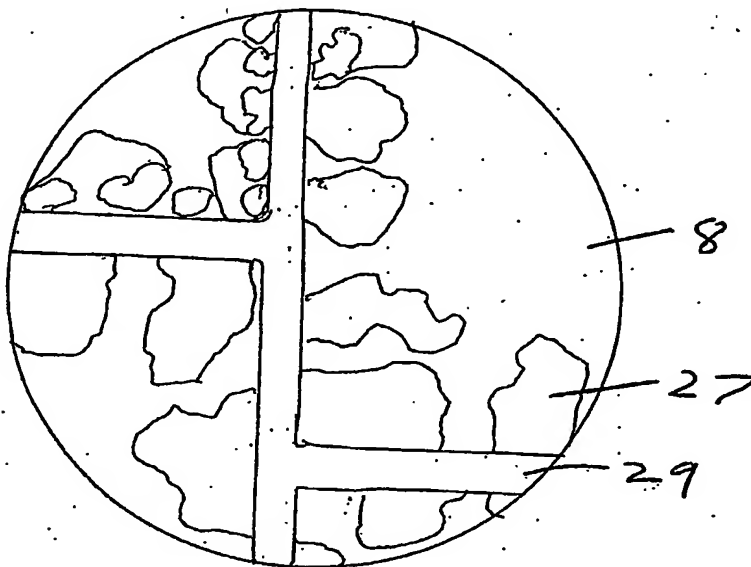
30
32

30
34

1C (c)

30

28

8

1C(d)

1·6

14

18

8

20

Figure 1C(e)

22

8

Figure 1C(f)

Figure 2B - Side view of
Anode with gradient surface area
and pore size

42 44 46

Figure 2A Side view diagrammatic
of Anode with high flow core

36

38

40

38

51

49

47

49

.52 .48

Fig. 3. R Anode with water

50

49

51

51

47

48

Fig. 3. A Anode with fuel media

54

58

56

60

63

62

71

72

74
76

68
69

61

64

86

78

4 , Diagrammatic representation of a fuel cell design with fuel

The stack case showing one possible way of orienting

106

104

112

110

108

liquid
exit

AIR
IN

AIR
OUT

IN
liquid

Figure 6 Sketch of manifolds placement relative to the stack case.

Figure 7c

146
136
132

Figure 7c Side view of a single
cathode paired with
a more massive anode.

134 134
132
136
132
132
144

Figure 7 b Side view of a flow
through anode
surrounded by two
cathodes.

138 140 142
136
132
134 135
132 134 136
132

Figure 7A Side view rarified
core anode surrounded
by two cathodes

160 162 164 168 170 172 174 175 176 178 180 166 170

Fig. 61. Diagram of separated anodes and cathodes
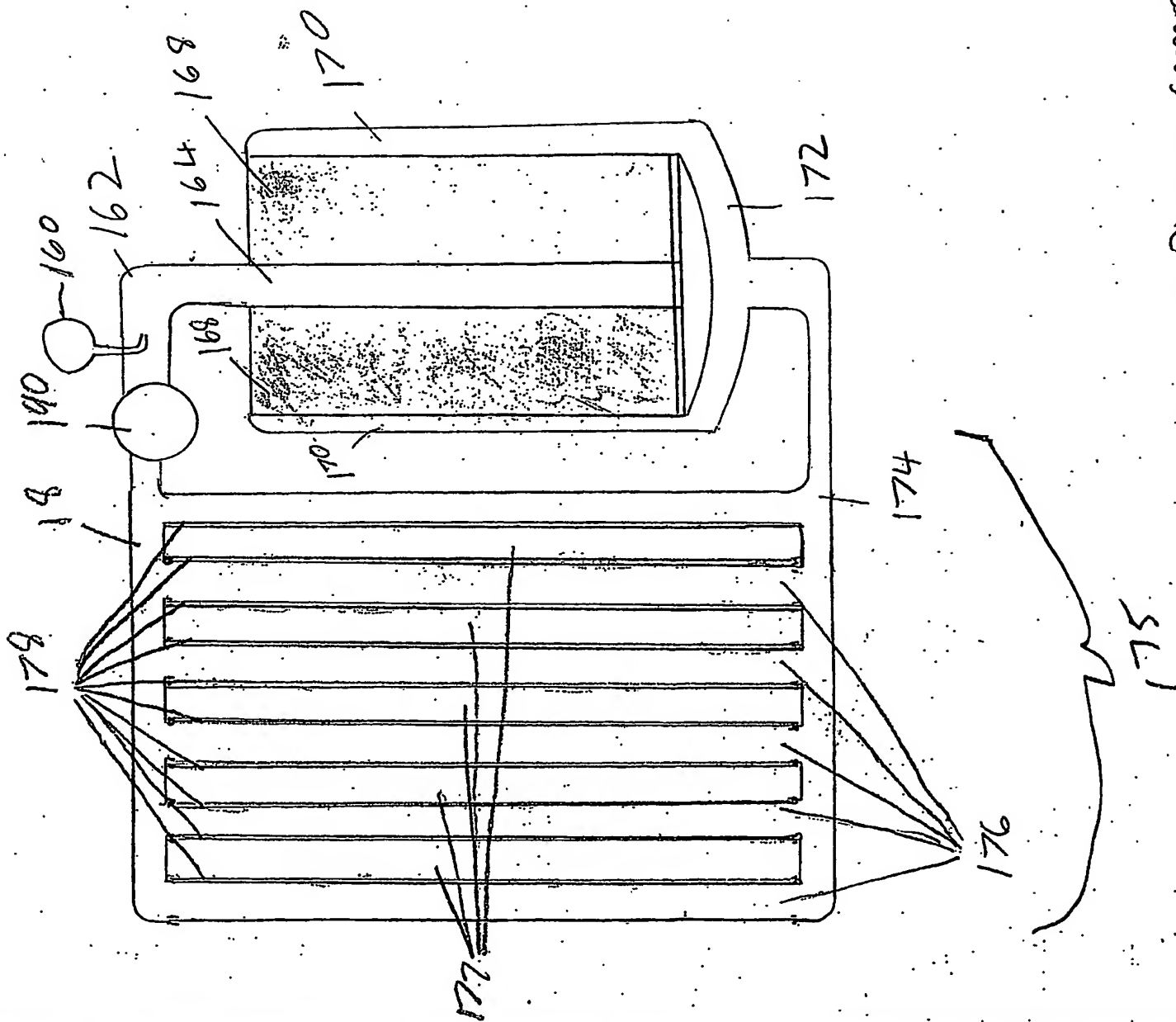
Flow through anode —

Flow by cathode —

Flow through anode

pump

148

150

158

154

152

151

157

160

153

155

156

Figure 8

Diagram of separated anodes and cathodes,
permutation demonstrating differential
number of anodes vs. cathodes and different

192

196

Stack

194

Fan

198
G

200

206

204

202

H_2O
electrolite
Fuel

148

A_sO

$1x^y$

q_a

Figure 9 The water, thermal, pressure management system